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DEVELOPMENT OF CELLULOSE BASED PACKAGING MATERIAL FROM COFFEE PULP AND IMPROVEMENT OF ITS ANTIOXIDANT CAPACITY

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ABSTRACT

A biodegradable packaging film was developed by substituting methylcellulose obtained from coffee pulp which offer favorable environmental advantages of recyclability and reutilization compared to conventional petroleum-based synthetic polymers. The films developed was optimized on the basis of thickness and tensile properties. With increase of ratio of substitution of natural methylcellulose with coffee pulp methylcellulose, thickness of the developed packaging material was decreasing. Significant difference in the thickness was observed when natural cellulose was substituted above 5% and below 20% with coffee pulp methylcellulose. Increasing concentration of coffee pulp methylcellulose resulted in decreased mechanical property, both tensile strength (TS) and elongation (%E). TS of 15% substituted ratio (42.81 N/m²) was found to be similar to control polypropylene (PP) films (42.83 N/m²) sample whereas there was a significant difference between %E of control with minimum substituted values. The optimized solution includes 85.19% of natural cellulose and 14.81% coffee pulp methyl cellulose for developing the film. Addition of 18.5g of antioxidants (extracted from coffee pulp) in the developed films resulted in more retention of Vitamin E Acetate (993.811 mg/l) compared to other samples thereby indicating a decrease in oxygen transmission rate.

KEYWORDS: Coffee Pulp, Biodegradable Films, Thickness, Tensile Characteristics, Antioxidant Capacity

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Original Article

1 INTRODUCTION

Coffee pulp (CP), identified as coffee fruit without seeds, or beans, is an abundant agricultural by-product, representing 43 % of the weight of the coffee fruit. The mucilage is a thin hydrated slimy layer which forms (5-14)% of the dry weight of the fruit (Coffee Board of India, 2011). Significant amount of parchment is also generated prior to roasting. On an average 1 ton of green coffee generates about 650kg of spent coffee (SC), and about 2kg of wet SC are obtained to each kg of soluble coffee produced (Pfluger, 1975).

In coffee producing countries, coffee wastes and by-products constitute a source of severe contamination and pose serious environmental problem. Coffee processing units that are located in almost each coffee estate pose threat to the environment because of unsafe disposal of coffee pulp, husk and effluents leading to pollution of water and land around the processing units.

Recently **solid state fermentation (SSF)** with coffee by-products, especially CP and CH has gained interest owing to its importance in recent developments in biomass conservation, in solid waste treatment and its application to produce secondary metabolites. Exploration using coffee by-products for the production of enzymes

like amylase, protease and xylanase was carried out using fungal organisms such as *N.crassa, A.oryzae, Penicillium spp.* and *A.niger* (Murthy and Naidu, 2010). In recent years, special emphasis has been given to solid state production of citric acid by fungal strains mainly *Aspergillus niger* (Kumar and Jain, 2008). Production of citric acid in basal CH medium by *A. niger* under solid state fermentation was reported by Ramachandra *et al*. (2013).Bhoite *et al*. (2013) studied the production of **gallic acid** through the transformation of CP tannins by *P.verrucosum.* Among the fungi isolated from coffee by-products, *P.verrucosum* produced 35.23 µg/g of gallic acid with CP as a sole carbon source in SSF.

Usha and Appaiah (2011) investigated the suitability of coffee cherry CH for the **production of bacterial cellulose (BC)** by Gluconacetobacter hansenii UAC09.A systematic study on **cultivation on mushrooms** like *L.edodes*, *Pleurotus species* and *Flammulina velutipes* using such residues as CH, leaves and spent ground either, individually or in mixture are reported (Murthy and Manonmani, 2008).

CHs and CP have been reported to be used to **feed** farm animals (Mazzafera, 2002). The CP has 12% protein content and its incorporation upto 20 % in cattle diet, 5% in poultry feed, 3% in bird and 16 % in pig feed has been recommended. Productive performance, rumen fermentation and oxidative status of sheep fed diets supplemented with CP (8% and 16%) ensiled with 5% molasses were evaluated (Salinas-Rios *et al*., 2015). The study concluded that a supplemented sheep diet with CP up to 16% CP did not affect their productive parameters but reduced oxidative stress.

CH and hulls contain a great amount of cellulose and hemicelluloses which makes it comparable to wood. Bekalo and Reinhardt (2010) showed that the CH-wood board showed great promise for use in structural and non-structural panel products based on superior flexural and internal bond properties. Their results collate potential for substituting wood upto 50% with CH in the **production of particle board** products.

Agro-industrial byproducts are good sources of **phenolic compounds**, and have been explored as sources of natural antioxidants (Fki and Allouche Nandsayadi, 2005). CP in the extraction of polyphenols has been explored (Sera *et al*., 2000). Ramirez-Coronel *et al*. (2004) found four major classes of polyphenols (viz., flavan-3-ols, hydroxycinnamic acids, flavonols and anthocyanidins) in Arabica CP. For instance, the phenolic compounds tentatively identified by high performance liquid chromatography (HPLC) in fresh CP by Ramirez-Martinez (1988) are: chlorogenic acid (5-caffeoylquinic acid) (42.2% of the total of identified phenolic compounds), epicatechin (21.6%), 3,4-dicaffeoylquinic acid, (5.7%), 3,5-dicaffeoylquinic acid (19.3%), 4,5-dicaffeoylquinic acid (4.4%), catechin (2.2%), rutin (2.1%), protocatechuic acid (1.6%) and ferulic acid (1.0%). Later on, Clifford and Ramirez-Martinez (1991) additionally identified 5-feruloylquinic acid in CP. Condensed tannins (proanthocyanidins) are also important constituents of the fresh CP (Clifford and RamírezMartínez, 1991). Their concentration increases along CP drying and is greater in yellow coffee varieties than in red ones (Colmenares*et al*., 1994).

Characterization of **anthocyanins**, polyphenols, and the biological properties of coffee skin/CP were recently investigated (Murthy and Naidu, 2012).Anthocyanins were analyzed by HPLC with photodiode array detection and electrospray ionization mass spectrometry. The anthocyanins from CP yielded 25 mg of monomeric anthocyanins per 100 g of fresh CP on a dry weight basis. The purified anthocyanin was identified as cyanindin-3-rutinoside and cyanidin-3-glucoside. Coffee anthocyanins have shown multiple biological effects resulting in effective α-glucosidase and α-amylase inhibitory activities. It was concluded that coffee skin/CP are potential sources of colorants and bioactive ingredients to be used in formulated foods (Murthy *et al*., 2012).

Fresh CP can be easily processed into various food commodities like **jam, juice, concentrate, jelly and flavouring** (Madahava*et al*., 2004). Adriane *et al*. (2003) reported that CP and husk can be used as substrates for *C.fimbriata* for aroma production using SSF. Twenty-one volatile compounds corresponding to higher alcohols, acetates, terpenes, aldehydes, and volatile acids were identified by gas chromatography (GC) when coffee byproducts obtained from semi-washed process was used as a substrate (Bonilla-Hermosa*et al*., 2014). *Hanseniaspora uvarum* showed the best fermentation performance with 12 % w/v of CP, 1g/l of yeast extract and 0.3 g/l of inoculums.

In recent years, development of bio-based packaging material has gained attention due to its renewable and biodegradable nature. The resulting environmental impact of the high consumption of plastic material in the food industry has encouraged special efforts from the packaging industry to develop biodegradable packaging materials.

Biopolymer-based packaging materials originated from naturally renewable resources such as polysaccharides, proteins, and lipids or combinations of those components offer favorable environmental advantages of recyclability and reutilization compared to conventional petroleum-based synthetic polymers. Biopolymer films and coatings may also serve as gas and solute barriers and complement other types of packaging by minimizing food quality deterioration and extending the shelf life of foods (Guilbert *et al*., 1997; Krochta and De Mulder-Johnston 1997; Debeaufort *et al*., 1998). Moreover, biopolymer-based films and coatings can act as efficient vehicles for incorporating various additives including antimicrobials, antioxidants, coloring agents, and nutrients (Ozdemir and Floros 2001; Han and Gennadios, 2005).These applications utilize only a fraction of available quantity and the methods were not technically very efficient. In view of the above background another application for waste utilization was look upon. This study was taken up to develop methyl-cellulose based packaging material from cellulose extracted from coffee pulp and improve the antioxidant capacity of the developed films.

2 MATERIALS AND METHODS

2.1 Purification of Coffee Pulp

The coffee pulp was provided by Regional Coffee Research Centre (RCRS), Thandikudi, Tamil Nadu, India. The water soluble extractives were removed by mixing 4.0 g of ground coffee pulp with 76.0 ml of distilled water for 24 hours. After 24 hours, it was filtered and mixed with 76.0 ml of NaOH (0.25 M). After 18 hours, the mixture was vacuum dried and filtered. It was then put in reflux for three successive portions of a mixture containing 20 % (v/v) of nitric acid in ethanol. After each hour the mixture was replaced. After refluxing, the mixture was filtered and washed with distilled water until the filtrate did not turn to pink when phenolphthalein and a drop of NaOH (0.05 M) were added to it. The treated pulp was then dried (105˚ C /3 hours) and ground in blender (Rodrigues Filho *et al*., 2000).

2.2 Extraction of Holocellulose

Five gram of dried coffee pulp was added to a 250 mL beaker, containing 100 ml of 4 % sodium hypochloite and 0.5 mL of concentrated acetic acid. The beaker was then covered with a watch glass and placed on a bath at 75 ˚C for 1 hour with occasional stirring. The same amount of reagents was added to the beaker every hour for 2 more hours, totalizing a digestion period of 3 hour. The system was cooled to 10 ˚C in ice water, and then filtered. The holocellulose obtained was washed six times with ice water and dried in an oven at 105 ˚C for 6 h (Viera *et al*., 2007).

2.3 Extraction of Alpha Cellulose Content

Three grams of holocellulose was put into a 250 ml Erlenmeyer flask and mixed with 100 mL of a 5% KOH solution in inert atmosphere, which was obtained by adding nitrogen gas to the mix during the first 10 min of extraction. The flask was put in a water bath at 25 ˚C for 2 h, with constant stirring. The mix was filtered in a sintered crucible, and washed with 50 ml of a 5% KOH solution, and thenwith 100 ml of distilled water. The filtrate was put into a 1 l Erlenmeyer flask and the precipitation was performed by adding a solution containing equal parts of acetic acid and ethanol. The precipitate was hemicellulose-A.

The fibrous residue retained on the sintered crucible was transferred to a 250 ml Erlenmeyer flask, and the same procedure that was performed to obtain hemicellulose-A was followed. However, a 24% KOH solution was used. The fibrous residue on the sintered crucible was washed with 25 ml of a 24%KOH solution, then with 25 ml of 10% acetic acid solution and finally with 100 ml of distilled water. The filtrate was recovered in a 1 l Erlenmeyer flask into which a solution containing equal amounts of acetic acid and ethanol was added. The precipitate was hemicellulose-B.

The fibrous residue at the end of the process described above was washed with distilled water until the filtrate pH was neutral. Then, it was washed with 50 ml of acetone and dried at 105 ˚C for 3 h to obtain alpha cellulose (Viera et al., 2007).

2.4 Methylation of Coffee Pulp Cellulose

One gram of alpha cellulose was mercerized using a 50% NaOH solution for one hour at room temperature. The excess of the NaOH solution was taken out and then 3 ml of dimethyl sulfate was added to sample. After addition of DMS, 9 ml of acetone was added and the mix was left in a water bath at 50˚C for 3 hour, with occasional stirring. At the end of the reaction, the material was neutralized by a 10% acetic acid solution, filtered and finally washed with acetone. The methylcellulose obtained was dried in an oven at 50˚C for 6 hour (Mansour et al., 1994).

2.5 Determination of Antioxidant Activity

2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging was evaluated according to the method of Yen and Chen (1995) with minor modification. Air dried and powdered samples (1g) were extracted with 10 ml of methanol which was soaked overnight in a shaking incubator at room temperature. The methanolic extracts were filtered using whatman filter paper. From that 0.1, 0.2, 0.3 ml of concentration was taken for analysis. Standard ascorbic acid was taken in 0.1, 0.2, 0.3 ml of concentration for analysis. 6 ml of 0.004 % of DPPH in 80% methanol is added to all test tubes. The test tubes were kept in incubation for 30 minute at room temperature in dark. The absorbance was then read against a blank at 517 nm and calculated using Eqn. 2.1.

$$\text{Antioxidant Activity } (\%) = \frac{\text{Absorbance (Abs)of blank-Absorbance(Abs)of sample}}{\text{Absorbance (Abs)of blank}} \text{ x } 10 \qquad (2.1)$$

2.6 Extraction of Antioxidant Compounds

Dried CP was sterilized at 121˚C for 20 minutes, powdered and extracted using Soxhlet apparatus. The sample weighing 100g was loaded into separate glass columns and extracted in 1:10 (w/v) of sample and solvent mixtures of

isopropanol and water (60:40 v/v) at 27˚C. The extracts were pooled and distilled in a rotary evaporator at 50˚C and stored at 4˚C (Sheng *et al*., 2010)[1].

2.7 Film Making Procedure

Three gram of methylcellulose, substituted with 5% to 25% with that of coffee pulp source, was first dissolved in 25 ml of hot water (80˚C) and then 75 ml of cold water (10˚C). The dispersion was kept under constant agitation until complete solubilization. 1 ml of PEG was added and the solution was homogenized with a homogenizer at 24,000 rev min^{-1} for 5 min. Antioxidants were added and again homogenized. The final solution was kept in a vacuum oven at 80˚C for about 5 hour in order to remove air bubbles or dissolved air. It was then spread on 20 X 20 cm glass plates to 0.5 mm thickness. The films were dried at 60˚C in hot air oven for 25 min and then at room temperature and 65±3% relative humidity for 2 days (Ayranci and Tunc, 2003).

2.8 Determination of Film Thickness

Film thickness was measured using a thickness tester (model no 547-401), to the nearest 2.5 µm (0.001mm) accuracy at 5 random positions around the films. The average value of the 5 was taken.

2.9 Determination of Tensile Strength (TS) and Elongation (E)

Universal testing machine (UTM) (Packtest KC-3000) was used to measure TS and %E at break as shown in Plate 3.9. The tests were carried out according to ASTM D-882 standard test (ASTM, 1980).

The films were cut into strips of size 100 X 15 mm with a single edged razor blade. About 3 such specimen were prepared for each sample. The test sample was tightly clamped to the upper jaw and loosely clamped to the lower jaw. Thereafter tightly clamping the lower end of the specimen and the load was applied. With initial grip separation of 70 mm and cross head speed of 50 mm/min.

TS was calculated by dividing the maximum load for breaking film by cross sectional area and %E was calculated by dividing film elongation at rupture to initial gauge length, and the values were measured both in longitudinal and transverse directions to observe whether any difference in the orientation of polymer chain occurs.

2.10 Optimization of the Developed Packaging Material Based on Physical and Mechanical Properties

Optimization of the CP methylcellulose substitution ratio by comparing the physical and mechanical property of various substituted proportions of natural methylcellulose and CP methylcellulose was analyzed. The optimized ratio was used for improving thewater vapour transmission and antioxidant capacity of the developed films.

The optimization process was carried out using mixture design (simple lattice design) (Leardi, 2009) by Design Expert version 6.0.8 software (Stat-Ease Inc., Minneapolis, USA).The responses analyzed were thickness, TS and E at break. The optimization was done with specific criteria for each response keeping ratio of natural cellulose to be minimum and other responses as maximum. The optimization criteria is shown in Table 1.

Table 1: Optimization Criteria

Sl No	Parameter	Criteria	Value
1	Natural Cellulose	Minimum	-
2	CP Cellulose	Maximum	-
3	Tensile Strength	Target	42.83 N
4	Elongation	Maximum	-
5	Thickness	Target	0.036 mm

2.11 Determination of Antioxidant Capacity of Films

Five gram of almond oil was dispensed in each petridishes. They were then distributed among five groups for dark storage at 25˚ C and 57% RH during 30 days. One of them was the control group which was stored without film protection. Other two groups were made up by samples added with varying quantities of with or without additive. For these trials, the films were cut in circles with the same diameter of the petridishes and put on each oil sample to isolate it from the storage environment. The experiment was carried in triplicates (Perez *et al*., 2013).

Vitamin E acetate content was analysed by high performance liquid chromatography (HPLC) consisting of a LC-10ATVp pump, SCL10A system controller and a variable Shimadzu SPD-10AVp UV VIS detector and a loop injector with a loop size of 20 µl. The peak area was calculated with a CLASSVP software. Reverse-phase chromatographic analysis was carried out in isocratic conditions using a C-18 reverse phase column (250 x 4.6 mm i.d., particle size 5 µm, Luna 5 µ C-18; phenomenex, Torrance, CA, USA) at 25˚C. The mobile phase consisted of deionized water, methanol (HPLC grade) and acetic acid in the ratio of 85:15:1 at 1 ml/min flow rate (Franca *et al*., 2005). Thewavelength used was 210 nm. Vitamin E acetate standard solution was employed for the peak identification and quantification.

2.12 Statistical Analysis

All the experiments were determined in random order of triplicates. The statistical analysis of antioxidant capacity of films after antioxidants addition and physico-chemical parameters were performed by applying Analysis of Variance (ANOVA), Tukey test and Least Significance Difference test (LSD). The IBM SPSS software (SPSS 21.0, Chicago, IL, USA) was used for performing the statistical analysis. The effect of cellulose substitution on mechanical and barrier properties of films were investigated using mixture design (simple lattice design) (Leardi, 2009) by Design Expert version 6.0.8 software (Stat-Ease Inc., Minneapolis, USA).

3 RESULTS AND DISCUSSIONS

3.1 Determination of Antioxidant Activity of Coffee Pulp

The DPPH free radical scavenging activities of coffee pulp (CP) powder and ascorbic acid (AA) at various concentrations are presented in Figure 1. The addition of CP powder into the DPPH solution caused a rapid decrease in absorbance at 517 nm indicating the excellent scavenging capacity of the CP. The DPPH scavenging activities of CP was found to be less than that of AA.

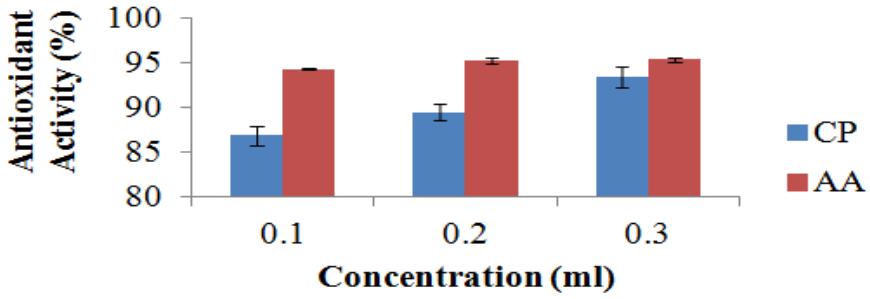

Figure 1: Antioxidant Activity of Coffee Pulp

3.2 Effect of Addition of Coffee Pulp Methylcellulose on Thickness

The effects of CP cellulose concentration on thickness of methylcellulose based films are shown in Figure 2. It was obvious that thickness was strongly influenced by the concentration of coffee pulp methylcellulose. Thickness of cellulose based films was inversely related to concentration of coffee pulp methylcellulose. As the concentration of coffee pulp methylcellulose increase, thickness at break significantly decreased. The highest value of thickness was observed with 5% substitution of CP cellulose (0.036mm), where as the lowest value was obtained with 25% substitution (0.023mm). All the values were compared with Poly propylene as control (0.036mm). There was no significance difference observed between 20 and 25% substitution of CP cellulose ($p>0.05$). It was observed that the control and 5% substituted film values were similar.

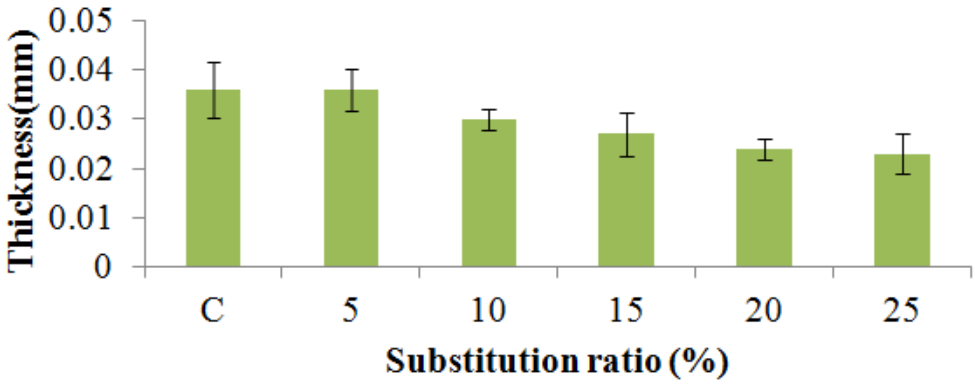

Figure 2: Changes in Thickness with Substitution of Coffee Pulp Methylcellulose

There was a different trend observed by Tongdeesoontorn *et al*. (2011). Study was conducted on the effect of increasing carboxymethyl cellulose (CMC) concentration on physical properties of bio-degradable cassava based films which showed that there was an increase in thickness with increase in CMC concentration. Increase in coffee pulpmethylcellulose concentration may lead to weakening of anti symmetric vibrations of carbon and oxygen bonds, probably due to formation of hydrogen bonds between carboxylic groups caused by added starch (Tong *et al*., 2008). Addition of coffee pulpmethylcellulose decreased water absorption bonds which may be due to O-H stretching and intermolecular/ intramolecular hydrogen bonds (Jiang *et al*., 2008).

3.3 Effect of Addition of Coffee Pulp Methylcellulose on Tensile Strength and Elongation

Effects of coffee pulp methylcellulose concentration on TS and E rate of methylcellulose based films are shown in Figure 3 and 4. Both the parameters were strongly influenced by the concentration of coffee pulp methylcellulose. TS and E at break of methylcellulose based films were inversely related to methylcellulose concentration. As the concentration of coffee pulp methylcellulose increased, both TS and E at break significantly decreased. The highest value of TS was observed with 5% substitution of coffee pulp methylcellulose(47.67N/m²), where as the lowest value was obtained with 25% substitution (40.54N/m²). All the values were compared with polypropylene (PP) as control (42.83N/m²). The highest value of E was observed with 5% substitution of coffee pulp methylcellulose(11.02%), whereas the lowest value was obtained with 25% substitution (8.74%). The E of PP, the control, was 17.02%. There was no significant difference observed between 20 and 25% substitution of coffee pulp methylcellulose.

A different trend was noticed by Tongdeesoontorn *et al*. (2011), in a study conducted on the effect of increasing carboxymethyl cellulose (CMC) concentration on physical properties of bio-degradable cassava based films. Their result showed that as the concentration of CMC increased, the TS of films significantly increased but the E at break significantly decreased. Both the parameters were found to be inversely related.

Gennadios *et al*. (1993) reported that films with higher E values usually required a lower load to cause film breakage. A decreased E values with increased ratio of substitution could be related to structural modification of the methylcellulose network by water which causes a greater flexibility in polymer structure. Similar results were obtained on the effect of humidity on E of blended chitosan- methyl cellulose films (Rachtanapoon and Wongchaiya, 2012).

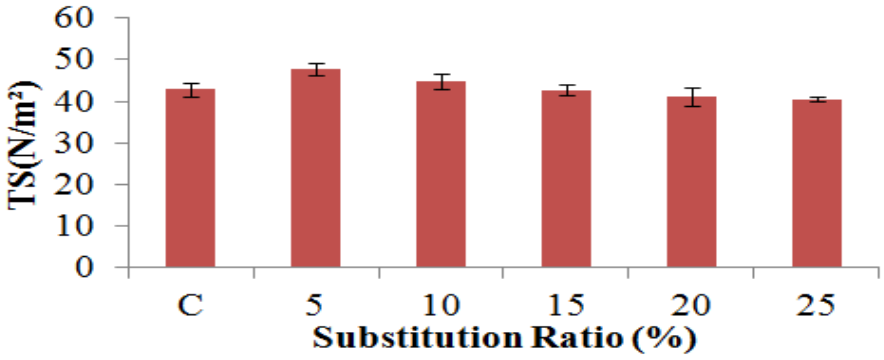

Figure 3: Changes in Tensile Strength with Substitution of Coffee Pulp Methylcellulose

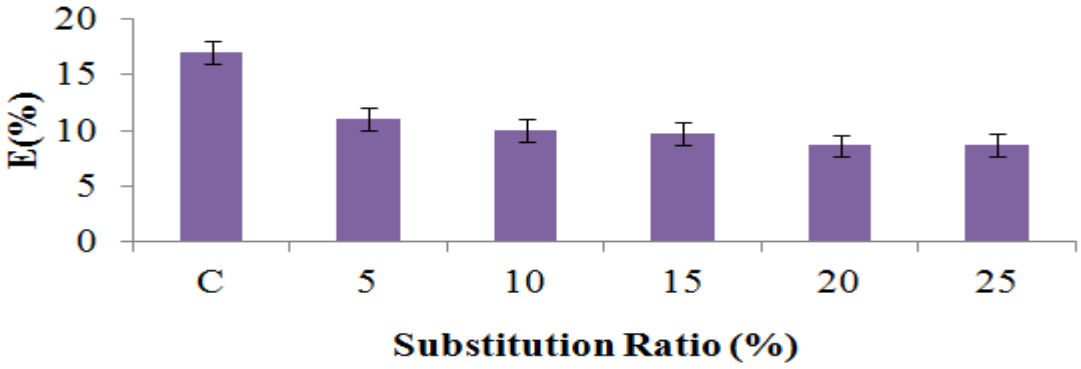

Figure 4: Changes in Elongation with Substitution of Coffee Pulp Methylcellulose

The decreasing TS of the cellulose based films with increasing concentration of coffee pulp methylcellulose is likely due to the weakening of inter molecular interaction between the hydroxyl group of starch and methyl group of cellulose (Li *et al*., 2008). During the processing and drying of the composite films, the original hydrogen bonds formed between starch molecules that do not be replaced by new hydrogen bonds between the hydroxyl group in starch molecules and hydroxyl and methyl groups in cellulose (Almasi *et al*., 2010). Absence of inter molecular interaction resulted in decrease of TS.

3.4 Optimization Results

The experimental results showed that substituting the film composition with coffee pulp methyl cellulose had a significant effect on the thickness as well as tensile properties of the developed film. With the increase in concentration of substituted coffee pulp methyl cellulose, there was a significant decrease in thickness, tensile strength and elongation. Based on the desired criteria, one optimized solution containing maximum substituted coffee pulp methylcellulose was found which include 85.19% of natural cellulose and 14.81% CP cellulose for developing the film.

3.5 Effect of Antioxidants Addition on Antioxidant Capacity of Films

Almond oil is a valuable functional ingredient due to its high polyunsaturated fatty acid content, which also makes it highly susceptible to oxidation. Tocopherols are the natural AO found in almond oil. The shelf-life of tocopherols can determine the start of the early signs of oxidative oil spoilage (lipid peroxides).

Data obtained from high performance liquid chromatography (HPLC) test in five samples were shown in Figure 5. The highest values were obtained for samples covered with PP (1003.43 mg/l) whereas lowest value was obtained for samples covered with optimized film without AO. With the increase of addition of AO, there was an increase of Vitamin E content. But there was no change with increase beyond 18 g.

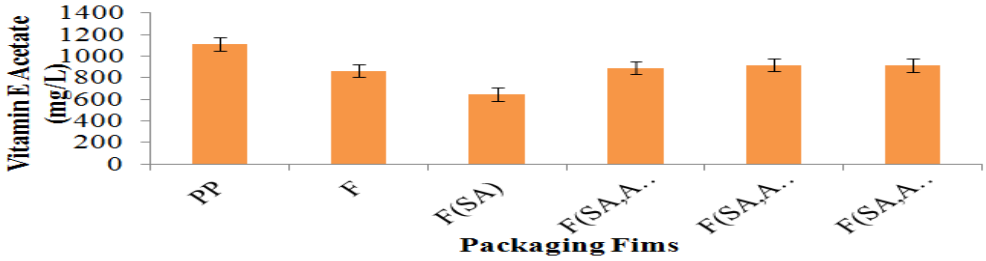

Figure 5: Changes in Vitamin E Acetate Content with Different film Composition

Perez *et al*. (2013) reported that film containing AO and based on high methoxyl pectin (HMP) showed a higher antioxidant activity upto 40 days of storage compared to the HMP and methyl cellulose (MC) film with AO.

The reason behind no effect beyond 18 g may due to the network constituted by natural and coffee pulp methylcellulose blend which resulted in a lower AO availability to exert its AO effect.

4. CONCLUSIONS

A biodegradable packaging film was developed by substituting cellulose obtained from coffee pulp which offer favorable environmental advantages of recyclability and reutilization compared to conventional petroleum-based synthetic polymers. The optimized solution based on physical and mechanical property includes 85.19% of natural cellulose and 14.81% coffee pulp methylcellulosefor developing the film.The characteristics of the developed film are comparable to

other edible films. Addition of coffee pulpAO in the developed films helped to improve the antioxidant capacity of the developed film. The new film products may exploit the utilization of coffee pulp cellulose.

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